On the Phase Shift Accompanying Reflection of Light from Evaporated Films and the Change of Effective Refractive Index as Function of the Thickness of Deposit

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journal or publication title	Science reports of the Research Institutes, Tohoku University. Ser. A, Physics, chemistry and metallurgy
volume	3
page range	740-747
year	1951
URL	http://hdl.handle.net/10097/26477

On the Phase Shift Accompanying Reflection of Light from Evaporated Films and the Change of Effective Refractive Index as Function of the Thickness of Deposit

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(Received October 25, 1951)

Synopsis

For the calculation of phase shift accompanying reflection from a thin film on base medium, the author used the surface of hypothetical single medium with an effective refractive index for the surface of lamella with one layer on base medium and expanded this method into the case of reflection from multiple layers on base medium. The phase shift and effective refractive index as function of the evaporated thickness were calculated in the limited cases of interest in optical practice, and if the dielectric deposit is evaporated and this lamella has the thickness with minimum or maximum resultant reflectivity, the following relations will be established at these thicknesses:

I) n term of effective refractive index turns to minimum or maximum value, respectively,

II) k term of effective refractive index turns to zero in all cases,

III) the curve of phase shift corresponds to the point of inflexion. These relations also hold true in multiple dielectric layers on a base medium. The phase shifts of air/aluminium, air/[aluminium(opaque) + ZnS], air/[aluminium (opaque) + $MgF_2(\lambda_0/4)$ +ZnS] interfaces were observed as a function of the thickness of final film from the displacement of Fizeau fringe.

I. Introduction

Several papers have considered the problem of the reflection and transmission of multiple films in its most general form. Weinstein(1) has dealt with this problem by the consideration of the electromagnetic disturbance in each medium and gave expressions for the reflectivity and transmissivity in terms of matrics. Polster(2) has found a reduction formula determining the reflection from a multilayer filter. Using Stoke's method,(3) Crook(4) has considered the expressions for the reflected and transmitted amplitudes of up to three films and formed a generalization for an arbitrary number of films. Vasicek(5) has dealt with the problem of reflection from multiple films using his idea of "basic amplitudes" and Wilcock(6) made a remark upon Vasicek method. The author(7) has induced the equation for calculating

(1) W. Weinstein, J. Opt. Soc. Am. **37** (1947), 576.
(2) H. D. Polster, J. Opt. Soc. Am. **39** (1949), 1038.
(3) R. W. Wood, *Physical Optics* (3rd edition) (1934), 193.
(4) A. W. Crook, J. Opt. Soc. Am. **38** (1948), 954.
(5) A. Vasicek, J. Opt. Soc. Am. **37** (1947), 623.
(6) W. L. Wilcock, J, Opt. Soc. Am. **39** (1949), 889.
(7) S. Nawata, Sci. Rep. RITU., A 1 (1951), 107.
S. Nawata, Jour. Phys. Soc. Japan. **7** (1952), 81, to be published.

the lamellar reflectivity with a single layer on a surface of metal by extending Nathanson and Bartberger's[(8)] equation for a layer on glass, and then, interpreting this equation as a reflectivity for a surface of hypothetical medium with an "effective refractive index", we calculated the reflectivity of multiple layers on a base medium. In this paper, we will calculate the phase shift accompanying reflection of light from evaporated films and the change of effective refractive index, as a function of the thickness of the deposit by the idea of hypothetical medium, and the phase shift will be measured.

II. Phase Shift and Effective Refractive Index

For the reflection of light from a thin film on base metal surface, as shown in Fig. 1 (a), if we can induce a single medium with an effective refractive index indicating as same reflectivity as this metal surface with a single layer, we can deal with this problem of two mediums as the one of surface reflection of single medium (see Fig. 1 (b)). Fresnel reflection coefficient of this surface is $(1-\bar{n})/(1+\bar{n}) = (1-n+ink)/(1+n-ink)$, using $\bar{n}$ for an effective refractive index.

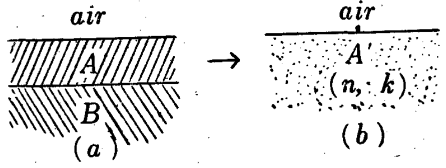

Fig. 1 Reduction of reflection from two mediums to reflection from one equivalent medium with an effective refractive index.

To know n an k, we put $(1-\bar{n})/(1+\bar{n}) = P-iQ$, then

$$n=(1-P^2-Q^2)/(P^2+Q^2+2P+1),\ k=2Q/(P^2+Q^2-1) \qquad (1)$$

The effective refractive index can be calculated from equation (1) and the phase shift due to reflection as $\Psi = tan^{-1}(Q/P)$. Obviously, the refractive index and phase shift are the functions of the thickness of deposit and of the optical constant of each medium.[(7)]

The same reduction of layer can be applied to two or more thin films. When two thin films A and B are on a base medium C as shown in Fig. 2, we induce the medium B' with an effective refractive index indicating same reflectivity as the surface of (B+C) mediums, assuming that B layer contacts with air directly, and moreover, we replace (A+B') mediums with one medium A'. Consequently, we can deal with the problem of three mediums as the one of a single medium with an effective refractive index. When multiple layers are on the base medium, we can deal with the resultant reflection by successively reducing the number of mediums, if the substitution of equivalent mediums is made upwards from the lowest medium.

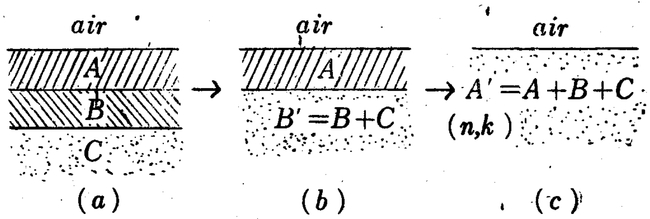

Fig. 2 Reduction of reflection from three mediums to reflection of one equivalent medium with an effective refractive index.

(8) J. B. Nathanson & C. L. Bartberger, J. Opt. Soc. Am. 29 (1939), 417.

It is known from the Fresnel coefficient that the reflection phase shift at air/glass interface is $\pm\pi$. To unify the treatment of metals and dielectrics, the negative sign is here adopted, the phase shift being interpreted as an advance.

Case I. Front Surface Deposit of Alminium on Glass.

The results calculated of phase shift and of effective refractive index of aluminium film on glass are shown in Fig. 3 as the function of the thickness of deposited aluminium along with the reflectivity calculated in the previous paper.[7]

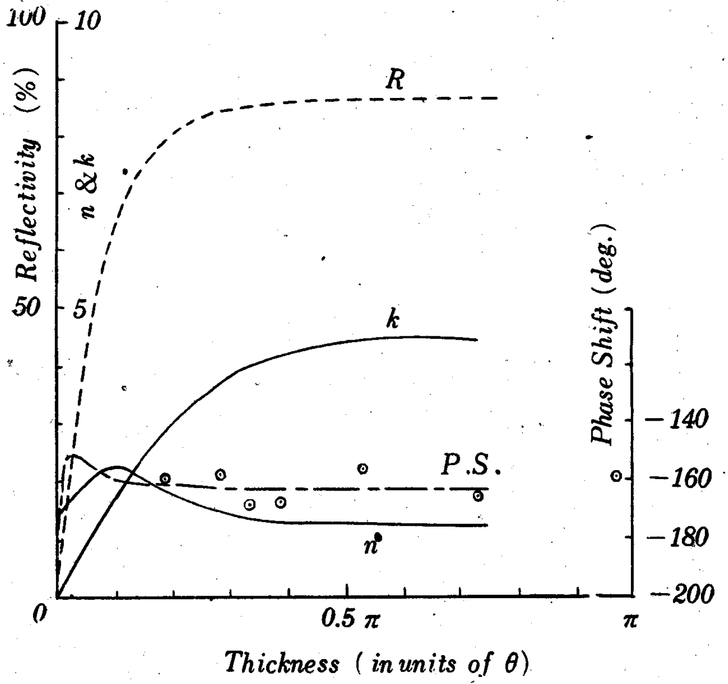

Fig. 3 Phase shift (◉ observed) and change of effective refractive index of the aluminium mirror along with the variation of reflectivity.

The optical constants (n, k) of aluminium for light of wavelength 5,890Å were taken as 1.36 and 4.45,[9] respectively. In the process of calculation, it was assumed that these constants did not change with thickness of deposit and that the film was continuous and uniform. When non-film was on glass, the phase shift indicated that of glass itself ($-\pi$), and the phase shift retarded with the increase of the thickness of aluminium film, advanced again to attain to the constant phase shift of opaque aluminium film.

The n term of refractive index started from the value of glass itself ($n=1.5$) and attained to the value of opaque aluminium ($n=1.36$) after passing a maximum value. The k term of refractive index starting from zero gradually came to the constant value of opaque aluminium ($k=4.45$).

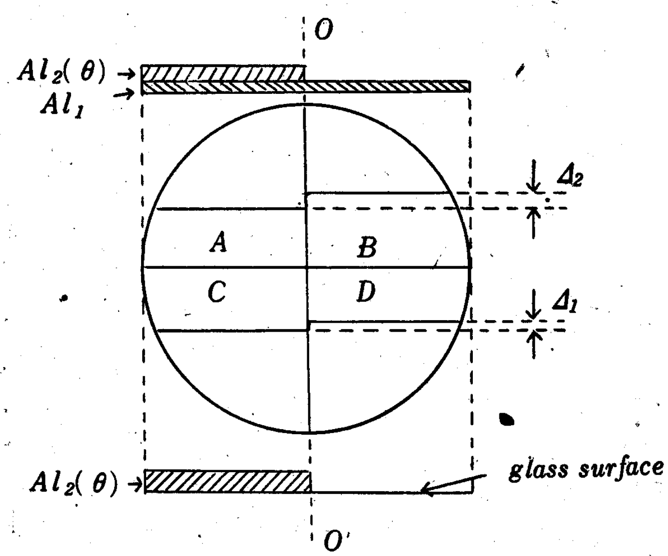

Fig. 4 Construction and fringe displacement of the test flat of an aluminium surface.

In order to observe the phase shift as a function of

(9) Landolt-Börnstein, *Physikalisch-chemische Table* 5, Auf. II, 906.

thickness of aluminium, we divided the surface of optical flat glass into four parts (A, B, C, D) (see Fig. 4), and the aluminium evaporated from tungsten heater at a pressure of about 5×10^{-5} mmHg, was deposited opaquely on the half (A+B) of flat glass. Next, the aluminium film with proper thickness was evaporated on the parts A and C. We set other optical flat with almost opaque aluminium film on the former flat to form an air-gap interferometer. We adjusted the interferometer and made the Fizeau fringes perpendicular to the boundary O–O′. The interferometric observations were carried out with a low power measuring microscope and we calculated the thickness of aluminium film on the part C from the fringe displacement (Δ_2) of boundary of the parts A and B, then we observed the fringe displacement (Δ_1) between C and D parts. Using these Δ_1 and Δ_2, we measured the reflection phase shift Ψ_{al} of air/aluminium interface as the function of deposited alumidium thickness from the following equation $\Psi_{al}=-\pi-\Delta_1+\Delta_2$, where Δ_1 and Δ_2 should be used as the absolute quantity regardless of the orientation of displacement. The result observed are shown in Fig. 3.

Case II. Dielectric deposits on opaque aluminium layer.

The results calculated and observed of the mirror consisting of [aluminium (opaque)+ZnS] are shown in Fig. 5 as the function of the thickness of ZnS. The optical constants of aluminium were used as $n=1.36$ & $k=4.45$ and that of ZnS was used as $n=2.4$ for light of wave length 5,890Å.

N term of the effective refractive index of this resultant layer starting from the value of aluminium itself, decreased as ZnS film grew thicker and came to the minimum value when the deposited thickness of ZnS was $\theta\fallingdotseq0.75\pi$, and to the maximnm value when $\theta\fallingdotseq1.8\pi$. The former thickness corresponded to that with minimum reflectivity and the latter to that with maximum reflectivity. The value of n returned to initial value when the deposit thickness of ZnS was 2π.

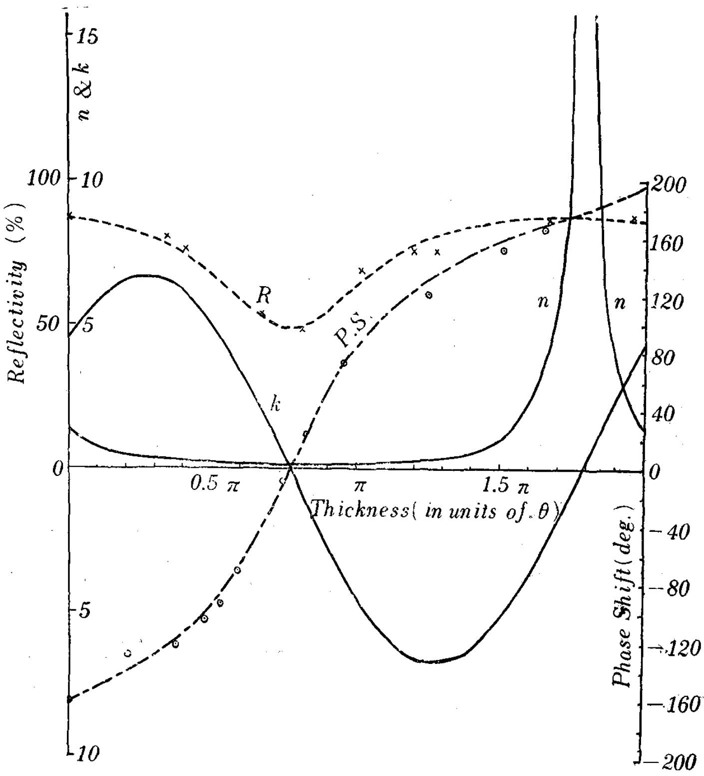

Fig. 5 Phase shift (⊙ observed) and change of effecitve refractive iedex, of an aluminium surface covered with ZnS layer along with the variation of reflectivity (× observed).

The change of k term, starting from the value of aluminium itself returned to initial value at the thickness of 2π. In this period, k became zero two times, one of which corresponded to the thickness of ZnS with minimum reflectivity and the other to that with maximum reflectivity.

The curve of the phase shift starting from the value of aluminium itself, retarded with the increase of the thickness of ZnS, and the curve had a point of inflexion at the thicknesses with minimum and maximum reflectivities.

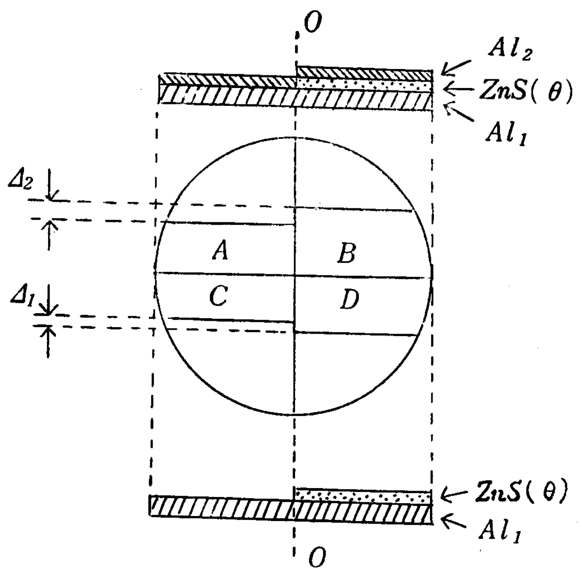

Fig. 6 Construction and fringe displacement of the test flat consisting of [Al(opaque) + ZnS(θ)].

The observation of phase shift was made as follows: the surface of an optical glass flat was divided into four parts (A, B, C, D) and the aluminium was evaporated opaquely on all four parts (see Fig. 6). Next, ZnS dielectric film with proper thickness was evaporated on the parts B and D. Finally, the opaque aluminium film was again evaporated on the parts A and B. Then, we formed an air-gap interferometer and made the Fizeau fringes perpendicular to the doundary O–O′ (see Photo. 1). We calculated the thickness of ZnS dielectric layer on the parts B and D, from the fringe displacement (Δ_2) between the parts A and B, and observed (Δ_1) between the parts C and D. Using these Δ_1 and Δ_2, we obtained the reflection phase shift of air/[aluminium +ZnS(θ)] interface from the equation $\Psi = \Psi_{al} + \Delta_1 + \Delta_2$, where Δ_1, Δ_2 were used as the absolute quantity regardless of the orientation of fringe displacement, and the mean value observed in former case as the phase shift of opaque aluminium itself was used as Ψ_{al}.

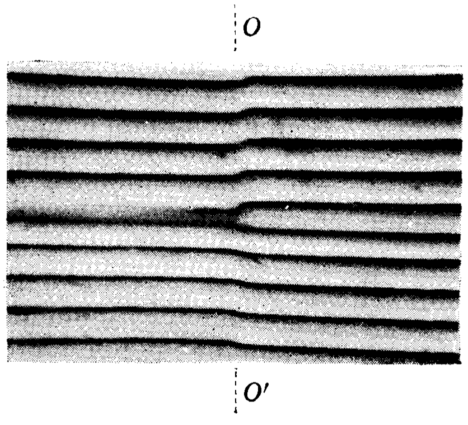

Photo. 1
Fizeau fringe of the test flat consisting of [Al(opaque) + ZnS(θ)].

Case III. Dielectric Deposits on Glass

The phase shifts and the change of effective refractive indexes of MgF_2 ($n=1.3$) and ZnS ($n=2.4$) deposits on glass ($n=1.5$) were calculated as a function of the thickness of deposited layer and are shown in Fig. 7 and Fig. 8 along with the variation of the reflectivity.

The phase shift indicated that of glass itself ($-\pi$) when non-film was on glass, retarded first with the increase of the thickness of MgF_2 layer, but advanced abruptly from about ($\theta \fallingdotseq 0.95\pi$) and it came to ($-\pi$) when the thickness of layer was $\lambda_0/4(\theta=\pi)$, and this thickness corresponded to the point of inflexion of the curve of phase shift. The change after this was reverse to that in the range θ 0 to π.

The term n starting from that of glass itself (n=1.5) returned again to the intial value at $\theta=2\pi$ after passing a minimum value. The thickness of deposit with this minimum value corresponded to that with minimum reflectivity.

The term k which was zero when non-film was on glass, had the possitive sign in the range θ 0 to π, and the sign of k in the range θ π to 2π was negative.

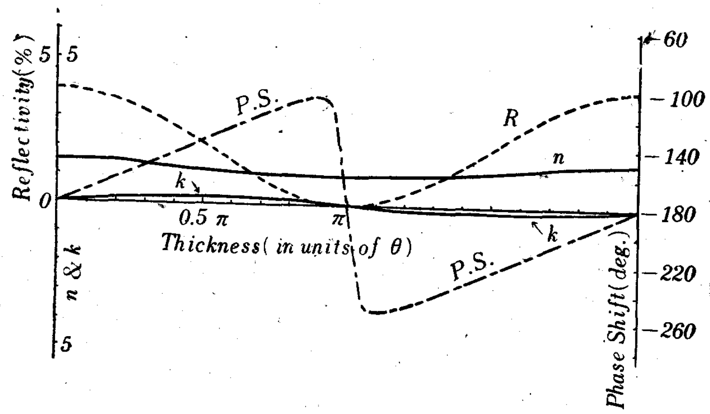

Fig. 7 Phase shift and change of effective refractive index of the glass with MgF_2 layer along with the variation of reflectivity.

When ZnS dielectric layer was evaporated on glass, the phase shift and change of terms n and k were in the opposite tendency to those of MgF_2 dielectric layer on glass as shown in Fig. 8. In this case, k was zero and the phase shift was ($-\pi$) at the thickness of deposit in which n was maximum. This thickness corresponded to that with maximum reflectivity and to the point of inflexion of the curve of phase shift.

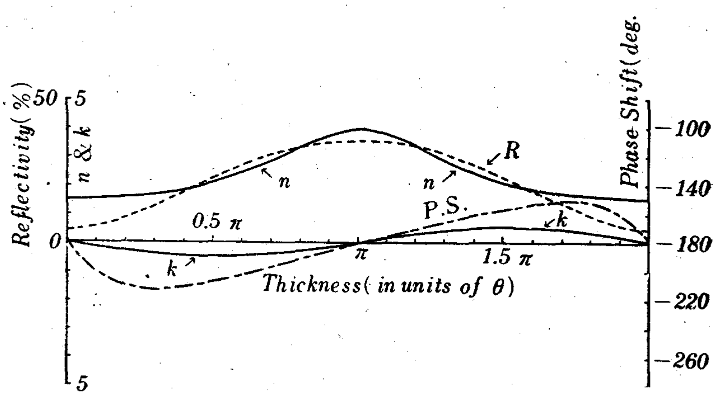

Fig. 8 Phase shift and chage of effective refractive index of the glass with ZnS layer along with variation of reflectivity.

No observations were made on the phase shifts of dielectric layers deposited on glass, because the width of Fizeau fringe became too broad in these cases to get the accuracy desired.

From the results mentioned above, we obtained the following synthetic expression in which the term n was minimum or maximum respectively, the term k was always zero and the curve of phase shift had a point of inflexion at these thicknesses when the dielectric deposit was evaporated and the lamella had minimum or maximum reflectivity. These relations also held true in multiple dielectric layers on a base medium.

III. The Mirror of [Al(opaque)+$MgF_2(\lambda_0/4)$+ZnS]

The phase shift and the change of refractive index calculated in multiple layers which consists of [Al(opaque)+$MgF_2(\lambda_0/4)$+ZnS] are shown in Fig. 9 along with the variation of the reflectivity. The synthetic expressions in refractive index and phase shift mentioned above held true also similarly in this multiple mirror.

The test flat used in the observation of phase shift was made as follows: the surface of optical glass flat was divided into six parts A, B, C, D, E, F (see Fig. 10) and aluminium was deposited opaquely on all six parts. Next, MgF_2 with $\lambda_0/4$ was deposited on the part B, C, E, F and ZnS on the parts C, F with proper thickness. Finally, opaque aluminium was deposited again on the parts A, B, C. Consequently, each part of flat was constructed as follows:

A part: Al_1+Al_2,
B part: $Al_1+MgF_2(\lambda_0/4)+Al_2$,
C part: $Al_1+MgF_2(\lambda_0/4)+ZnS(\theta)+Al_2$,
D part: Al_1 only,
E part: $Al_1+MgF_2(\lambda_0/4)$,
F part: $Al_1+MgF_2(\lambda_0/4)+ZnS(\theta)$.

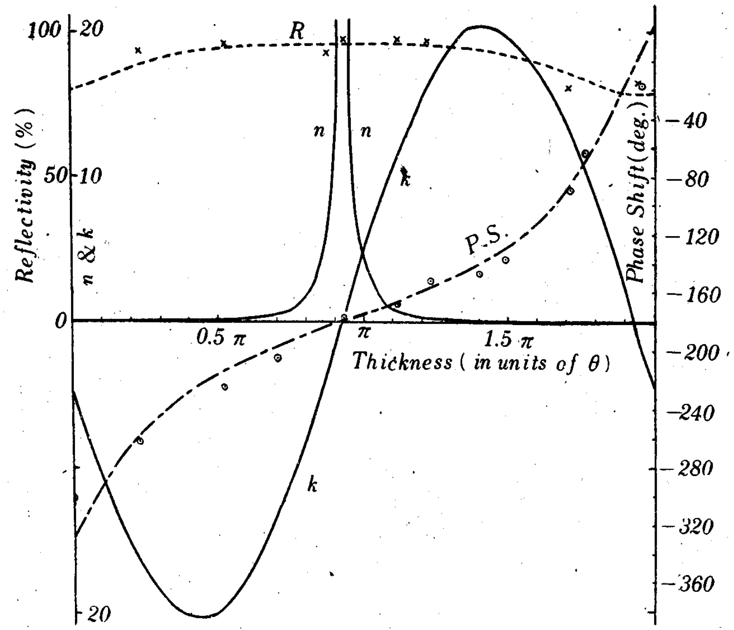

Fig. 9 Phase shift (◉ observed) and change of effective refractive index of the mirror consisting of [Al(opaque) $+MgF_2(\lambda_0/4)+ZnS$] along with the variation of reflectivity (× observed).

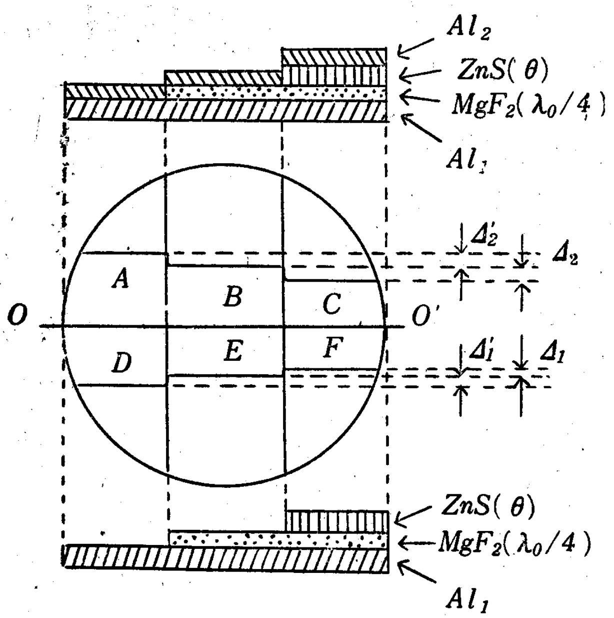

Fig. 10 Construction and fringe displacement of the test flat consisting of [Al(opaque) $+MgF_2(\lambda_0/4)+ZnS(\theta)$].

We made Fizeau fringes parallel to the boundary O–O′ and observed the fringe displacement Δ_1, Δ_2, Δ_1', Δ_2' of the boundaries of the parts (E/F), (B/C), (D/E), (A/B), respectively. The thickness of $MgF_2(\lambda_0/4)$ should be evaporated accurately by observing the fringe displacement Δ_2', and the phase shift Ψ of the mirror consisting of [Al(opaque)$+MgF_2(\lambda_0/4)+ZnS(\theta)$] from the following equation: $\Psi=\Psi_{al}+(\Delta_1+\Delta_1')+(\Delta_2+\Delta_2')$, where the result observed in case (I) was used as Ψ_{al}, and the thickness of ZnS was calculated from the fringe displacement Δ_2. The results observed are plotted in Fig. 9.

Summery

For the calculation of phase shift accompanying reflection from thin film on base metal surface, the author used the surface of single medium with an effective refractive index for the surface mentioned above and expanded this method on the case of multiple layers on base medium. We indicated the results calculated the changes of phase shift and effective refractive index as function of the evaporated thickness and discussed the relations between these quantities and lamellar reflectivity. In some cases, the phase shift were observed.

In conclusion, the author expresses his hearty thanks to Mr. K. Takashima for his assistance during the experiments and to Miss K. Yoshida for her earnest help during the numerical calculations.

The present investigation has been supported partly by the Grant in Aid for Fundamental Scientific Research of the Ministry of Education (No. 4064).